

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of	:	Attorney Docket No. 2006_1228A
Ya XU et al.	:	<b>Confirmation No. 6659</b>
Serial No. 10/587,940	:	Group Art Unit 1734
Filed September 29, 2006	:	Examiner Sarah Van Oudenaren
INTERMETALLIC COMPOUND Ni3Al CATALYST FOR REFORMING METHANOL AND METHANOL REFORMING METHOD USING SAME	:	<b>Mail Stop: AMENDMENT</b>

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**DECLARATION UNDER 37 C.F.R. § 1.132**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Ya Xu, the undersigned, a citizen of China, residing in Tsukuba-shi, Japan, do hereby declare:

**1. Background Information**

- I am a co-inventor of the above-identified application, Serial No. 10/587,940.
- I am being compensated for my work related to the above-identified application.
- I graduated from the University of Science and Technology Beijing, China in 1984 (B.S.); I graduated from the University of Science and Technology Beijing, China in 1989 (M.S.); and I received a Ph.D. from the University of Tsukuba, Japan in 1997.
- I was an assistant engineer in the Technology Division of Beijing Coal Machinery Co., Ltd. in China from 1984 to 1986; I was an engineer in the Consulting Division of China Metallurgical Enterprise Management Association from 1989 to 1993; I was a Domestic Research Fellow of JST, Tsukuba Magnet Laboratory, National Research Institute for Metals (NRIM) in Japan from 1997 to 2000 (NRIM was reorganized and merged into the National Institute for Materials Science (NIMS), the Assignee of the above-application, in 2001); and I was a NEDO researcher at Smart Structure Research Center, National Institute of Advanced

Industrial Science and Technology, in Japan from 2000 to 2003 (NEDO is New Energy and Industrial Technology Development Organization, a subsidiary of the Ministry of Economy, Trade and Industry).

6. At the present time, I am a senior researcher in the Alloy Catalyst Materials Group, Hydrogen Materials Unit, of NIMS.

7. I have co-authored several published articles relating to the technology claimed in the above-identified application, including: (i) Chun et al., "Catalytic properties of  $\text{Ni}_3\text{Al}$  foils for methanol decomposition," Catalysis Letters, Vol. 106, Nos. 1-2, Jan. 2006, pp. 71-75; (ii) Xu et al., "Catalytic properties of alkali-leached  $\text{Ni}_3\text{Al}$  for hydrogen production from methanol," Intermetallics, 13 (2005) 151-155; (iii) Chun et al., "Spontaneous catalytic activation of  $\text{Ni}_3\text{Al}$  thin foils in methanol decomposition," J. of Catalysis, 243 (2006) 99-107; and (iv) Xu et al., "Catalytic Properties of  $\text{Ni}_3\text{Al}$  Intermetallics for Methanol Decomposition," Materials Transactions, Vol. 45, No. 11 (2004), pp. 3177-3179. Copies of these publications are enclosed.

8. I have reviewed the present application, the pending claims, the Office Action dated December 9, 2010, and the references cited therein.

9. I have, under my control and direction, conducted the following experiments. The particulars and results of the experiments are set forth below.

10. **The Catalytic Activity of a Reaction System is Unpredictable**

11. In general, in the field of catalytic reactions, a compound exhibits catalytic activity to a specific reaction system. When the reaction system differs, for example, when the starting materials differ, the same compound would not necessarily exhibit the same catalytic activity in a different reaction system. Therefore, the catalytic activity of a reaction system is not predictable.

12. Accordingly, there is no reasonable expectation that applying a compound which is not disclosed in a reference would exhibit catalytic activity in the reaction system of the reference. Similarly, there is no reasonable expectation that a compound that exhibits effective catalytic activity in a specific reaction system of a reference would also exhibit effective catalytic activity in a completely different reaction system of a different reference.

13. **Experimental**

14.  $\text{NiAl}_3$  would not work as the Ni and Al containing catalyst of the Takuya et al. reference in view of the following experimental results.

15. A mixture of Ni and Al was produced in accordance with the method for preparing a mixture of Ni and Al disclosed in Takuya et al. (hereinafter referred to as "the Takuya et al. specimen"). In addition, a Ni<sub>3</sub>Al cold-rolled foil specimen was produced. Each specimen was then evaluated for its catalytic activity in a steam reforming reaction of a hydrocarbon, as disclosed in the Lessing reference.

16. The evaluation was conducted under the following feed conditions: CH<sub>4</sub>: 18 ml/min; H<sub>2</sub>O: 0.05 ml/min; and N<sub>2</sub>: 30 ml/min.

17. The Ni<sub>3</sub>Al cold-rolled foil specimen demonstrated an H<sub>2</sub> generation rate of 8400 (ml/min/m<sup>2</sup>) at 900°C, and this confirmed that the specimen has high catalytic activity. In the temperature region of 650 to 850°C, the H<sub>2</sub> generation rate was lower than the H<sub>2</sub> generation rate at 900°C, and this confirmed a lower catalytic activity in this temperature region.

18. The Takuya et al. specimen was evaluated under the same conditions as the Ni<sub>3</sub>Al cold-rolled foil specimen. The H<sub>2</sub> generation rate was 1200 (ml/min/m<sup>2</sup>) at 900°C. This confirmed that the catalytic activity was remarkably lower than the Ni<sub>3</sub>Al cold-rolled foil specimen. In addition, in the temperature region of 650 to 850°C, the H<sub>2</sub> generation rate of the Takuya et al. specimen was lower than the H<sub>2</sub> generation rate at 900°C. This reduction was similar to the reduction demonstrated in the Ni<sub>3</sub>Al cold-rolled foil specimen at the lower temperature range.

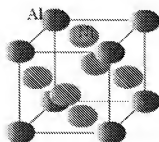
19. Thus, the Takuya et al. specimen demonstrated low catalytic activity in a steam reforming reaction of a hydrocarbon. These results show that the Takuya et al. specimen does not have the same catalytic activity in the steam reforming reaction of a hydrocarbon as it has in the methanol reforming reaction.

20. Accordingly, there is no reasonable expectation of success that applying the Ni<sub>3</sub>Al compound of Lessing, which demonstrates catalytic activity in a steam reforming reaction of a hydrocarbon, to the methanol reforming reaction of Takuya et al., would have the same catalytic activity in the Takuya et al. reaction as it has in the Lessing reaction.

21. **The Crystal Structure of Ni-Al Alloys**

22. In addition, Ni<sub>3</sub>Al has a crystal structure in which the arrangement of Ni atoms and Al atoms is ordered, as shown below.

(Ordering structure(L1<sub>2</sub>))



The ordered crystal structure of Ni<sub>3</sub>Al

23. However, in Ni-Al alloys, other than a Ni<sub>3</sub>Al alloy, the Ni atoms and Al atoms are distributed irregularly and randomly, and they do not have an ordered structure like a Ni<sub>3</sub>Al alloy. This different crystal structure leads to different physical and chemical properties.

24. The Examples in the specification of the present application confirm that the catalytic properties of Ni<sub>3</sub>Al are different from the catalytic properties of other Ni-Al alloys, such as Raney Ni. Fig. 1 of the present application shows the catalytic activity of Ni<sub>3</sub>Al and Raney Ni (through hydrogen generation rates) for a methanol reforming reaction. At a temperature of not less than 300°C, Ni<sub>3</sub>Al has a higher hydrogen generation rate than Raney Ni. Therefore, the catalytic properties are not consistent for all Ni-Al alloys.

25. In addition, Fig. 4 and Fig. 5 of the present application show the catalytic properties of several Ni-Al alloys having different compositions for a methanol reforming reaction. Ni-22 at% Al and Ni-24 at% Al are each Ni<sub>3</sub>Al single-phase examples. Ni-18 at% Al is an example having two phases: Ni<sub>3</sub>Al and Ni. Pure Ni is a Ni single-phase example. The Ni<sub>3</sub>Al single-phase examples have high catalytic activity, as shown in Figs. 4 and 5. The presence or absence of Ni<sub>3</sub>Al changes the content of the composition. Accordingly, among the Ni-Al alloys, the catalytic activity varies with the change in the composition.

26. The catalytic activity of a Ni-Al alloy is not predictable based upon the catalytic activity of a specific Ni-Al alloy, because the catalytic activity of a Ni-Al alloy varies with changes in its composition (i.e., it changes based upon the presence or absence of Ni<sub>3</sub>Al). Therefore, there would have been no reason to predict that applying a Ni<sub>3</sub>Al to a methanol reforming reaction would result in a high catalytic activity with a reasonable expectation of success in view of the disclosures of the references cited in the Office Action.

I further declare that all statements made herein of my own knowledge are true and all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Date: September 5, 2011

Luya

Ya Xu